



**EUROPEAN PATENT APPLICATION**

Application number : **94301634.5**

Int. Cl.<sup>5</sup> : **C08L 83/08**

Date of filing : **08.03.94**

Priority : **19.03.93 JP 85733/93**

Date of publication of application :  
**21.09.94 Bulletin 94/38**

Designated Contracting States :  
**DE FR GB**

Applicant : **Shin-Etsu Chemical Co., Ltd.**  
**6-1, Otemachi 2-chome**  
**Chiyoda-ku Tokyo 100 (JP)**

Inventor : **Kishita, Hirofumi**  
**19-1, Isobe 3-chome**  
**Annaka-shi, Gunma-ken (JP)**  
Inventor : **Yamaguchi, Kouichi**  
**12-22, Yachiyo-cho 1-chome**  
**Takasaki-shi, Gunma-ken (JP)**

Inventor : **Matsuda, Takashi**  
**A-505, Kofu Apt.,**  
**791-4, Yanase**  
**Annaka-shi, Gunma-ken (JP)**  
Inventor : **Fukuda, Kenichi**  
**1044-1, Shimonjo-machi**  
**Takasaki-shi, Gunma-ken (JP)**  
Inventor : **Kobayashi, Nobuyuki**  
**5-41, Isobe 4-chome**  
**Annaka-shi, Gunma-ken (JP)**  
Inventor : **Sato, Shinichi**  
**5-5, Isobe 3-chome**  
**Annaka-shi, Gunma-ken (JP)**  
Inventor : **Kinami, Hitoshi**  
**10-25, Isobe 4-chome**  
**Annaka-shi, Gunma-ken (JP)**

Representative : **Stoner, Gerard Patrick et al**  
**MEWBURN ELLIS**  
**York House**  
**23 Kingsway**  
**London WC2B 6HP (GB)**

**Organopolysiloxane compositions.**

An organopolysiloxane composition comprising (A) a linear, alkenyl-containing, fluorinated organopolysiloxane, (B) a cyclic fluorinated organosiloxane as a tackifier, (C) an organohydrogen-polysiloxane having at least two hydrogen atoms attached to a silicon atom in a molecule, and (D) a platinum group metal catalyst has excellent properties inherent to fluorinated silicone rubber, and cures into products having adhesion and close contact to various materials including metals, glass and resins.

**EP 0 616 012 A1**

This specification relates to organopolysiloxane compositions and more particularly, to fluorinated organopolysiloxane compositions, their manufacture and use, e.g. as packing, sealing and coating materials in electric and electronic fields.

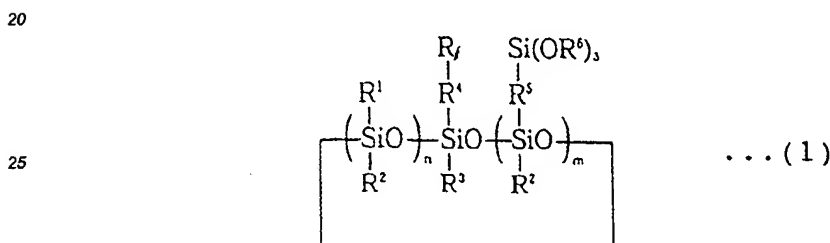
In addition to electric insulation, heat resistance, low-temperature resistance and weatherability inherent to silicone rubber, fluorinated silicone rubbers generally have improved surface properties including solvent resistance, water repellency, oil repellency and releasability, but suffer from poor adhesion to various support materials. There is a desire to have new fluorinated silicone rubbers available, and particularly having acceptable adhesion as well as good surface properties.

We have found that certain fluorinated cyclic organosiloxanes may exert a useful tackifying or adhesion-promoting effect in a composition of linear fluorinated organopolysiloxane having aliphatic-type unsaturation, typically together with organohydrogen polysiloxane for crosslinking. The compositions, this use of the cyclic organosiloxanes, and methods of preparing the compositions, are independent aspects of the disclosure.

A specific product proposal herein is an organopolysiloxane composition comprising

(A) a linear fluorinated organopolysiloxane having in a molecule at least two monovalent hydrocarbon groups each having an aliphatic unsaturated bond attached to a silicon atom, and further having at least one group selected from monovalent perfluoroalkyl and monovalent perfluoropolyether groups attached to a silicon atom,

(B) a cyclic fluorinated organosiloxane of the general formula (1):



wherein  $R^1$  is a substituted or unsubstituted monovalent hydrocarbon group having an aliphatic unsaturated bond,  $R^2$ ,  $R^3$  and  $R^6$  are independently selected from substituted or unsubstituted monovalent hydrocarbon groups free of an aliphatic unsaturated bond,  $R^4$  and  $R^5$  are independently selected from substituted or unsubstituted divalent hydrocarbon groups free of an aliphatic unsaturated bond,  $R_f$  is a monovalent perfluoropolyether group or monovalent perfluoroalkyl group, and letters  $m$  and  $n$  are numbers satisfying  $n + m = 3$ ,  $n \geq 1$ , and  $m \geq 1$ ,

Other components typically included are (C) an organohydrogenpolysiloxane having at least two hydrogen atoms attached to a silicon atom in a molecule, and (D) a platinum group metal catalyst.

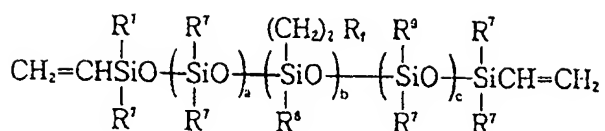
The composition is obtainable simply, e.g. by blending the above-mentioned components together, preferably adjusting the amount of the organohydrogenpolysiloxane such that there is 0.5 to 5 mol of SiH group per mol of the aliphatic unsaturated group in the entire composition. In our work, we have found that such compositions can have the good properties typical of fluorinated silicone rubber and also cure to a product having adhesion and close contact to various materials such as glass, aluminum, iron and epoxy resins. It seems that component (B) acts as an effective tackifier. The cured products were also fully resistant against solvent swelling.

#### BRIEF DESCRIPTION OF THE DRAWING

The only figure, FIG. 1 is a perspective view of an assembly for a shearing adhesion test.

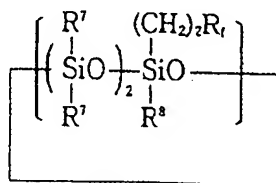
One component of the organopolysiloxane composition is a linear fluorinated organopolysiloxane having in a molecule at least two monovalent hydrocarbon groups each having an aliphatic unsaturated bond attached to a silicon atom. It further has at least one group selected from monovalent perfluoroalkyl and monovalent perfluoropolyether groups attached to a silicon atom. The unsaturated groups may terminate the Si chain.

Preferred examples of the linear, alkenyl group - containing, fluorinated organopolysiloxane include polymers of the following general formula (7).

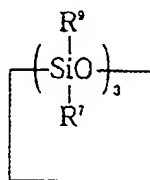


(7)

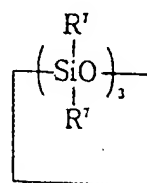
Such polymers are obtainable e.g. by copolymerizing a cyclotrisiloxane of formula (2), a cyclotrisiloxane of formula (3) and/or a cyclotrisiloxane of formula (4) with a compound of formula (5) in the presence of an acid or alkali catalyst. Alternatively, such polymers are obtainable by polymerizing a cyclotrisiloxane of formula (2), a cyclotrisiloxane of formula (3) and/or a cyclotrisiloxane of formula (4) in the presence of a silanolate catalyst of formula (6).



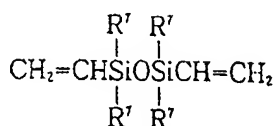
(2)



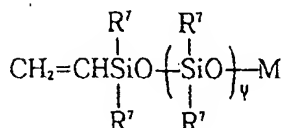
(3)



(4)



(5)

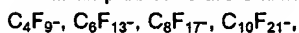


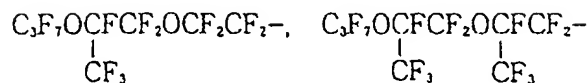
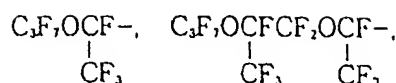
(6)

In formulae (2) to (7),  $\text{R}^7$  and  $\text{R}^8$  are independently selected from substituted or unsubstituted monovalent hydrocarbon groups having 1 to 12 carbon atoms free of an aliphatic unsaturated bond,  $\text{R}^9$  is a monovalent hydrocarbon group having 2 to 12 carbon atoms and having an aliphatic unsaturated bond, and  $\text{Rf}$  is a monovalent perfluoropolyether group or monovalent perfluoroalkyl group having 1 to 18, especially 1 to 12 carbon atoms.

Examples of the hydrocarbon group represented by  $\text{R}^7$  and  $\text{R}^8$  include alkyl groups such as methyl, ethyl, isopropyl and butyl, cycloalkyl groups such as cyclohexyl and cyclopentyl, aryl groups such as phenyl, tolyl and xylyl, aralkyl groups such as benzyl and phenylethyl. The presence of substituents (e.g. halo or cyano) is acceptable if not detrimental to the effect of the composition. Examples include halogenated hydrocarbon groups such as chloromethyl, chloropropyl, chlorocyclohexyl and 3,3,3-trifluoropropyl, and cyano hydrocarbon groups such as 2-cyanoethyl. Examples of the hydrocarbon group represented by  $\text{R}^9$  include vinyl, allyl and ethynyl groups.

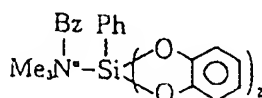
Examples of  $\text{Rf}$  are shown below.





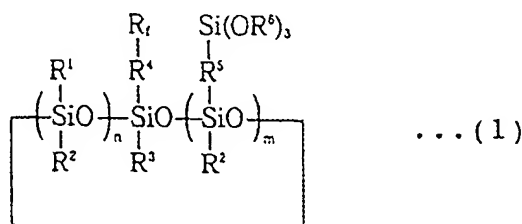
In formula (7), letters a, b and c are integers in the range: a = 2 to 3,000, b = 1 to 1,000, and c = 0 to 50.

Examples of the acid and alkali used as the polymerization catalyst include alkali hydroxides such as lithium hydroxide, sodium hydroxide, and potassium hydroxide, alkali siliconates such as lithium silicate, sodium silicate, and potassium silicate, quaternary salt hydroxides such as tetrabutylphosphine hydroxide and tetramethylammonium hydroxide, sulfuric acid, sulfonic acids such as trifluoromethane sulfonic acid, and five coordinate silicon compound of the following structural formula.



The fluorinated organopolysiloxane containing a monovalent hydrocarbon group having an aliphatic unsaturated bond of formula (7) which is prepared by a polymerization method as mentioned above preferably has a viscosity of 100 to 50,000 centistokes.

Another component is a cyclic fluorinated organosiloxane, serving as a tackifier or adhesion-promoter. Typically it accords with the general formula (1).

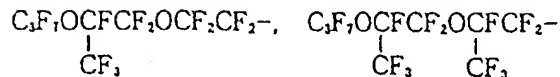
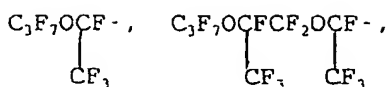


In the formula, R<sup>1</sup> is a substituted or unsubstituted monovalent hydrocarbon group having 2 to 12 carbon atoms and having an aliphatic unsaturated bond, R<sup>2</sup>, R<sup>3</sup> and R<sup>6</sup> are independently selected from substituted or unsubstituted monovalent hydrocarbon groups having 1 to 12 carbon atoms free of an aliphatic unsaturated bond, R<sup>4</sup> and R<sup>5</sup> are independently selected from substituted or unsubstituted divalent hydrocarbon groups having 1 to 12 carbon atoms free of an aliphatic unsaturated bond, R<sup>f</sup> is a monovalent perfluoropolyether group or monovalent perfluoroalkyl group having 1 to 18, especially 1 to 18 carbon atoms, and letters m and n are numbers satisfying n + m = 3, n ≥ 1, and m ≥ 1.

Examples of the hydrocarbon group represented by R<sup>1</sup> include vinyl, allyl, butenyl and styryl groups. Examples of the hydrocarbon group represented by R<sup>2</sup>, R<sup>3</sup> and R<sup>6</sup> include alkyl groups such as methyl, ethyl, isopropyl and butyl, cycloalkyl groups such as cyclohexyl and cyclopentyl, aryl groups such as phenyl, tolyl and xylyl, aralkyl groups such as benzyl and phenylethyl. The presence of substituents is acceptable if not detrimental to the effect of the composition, for example halo or cyano. Substituted possibilities include halogenated hydrocarbon groups such as chloromethyl, chloropropyl, chlorocyclohexyl and 3,3,3-trifluoropropyl, and cyano hydrocarbon groups such as 2-cyanoethyl. Examples of the divalent hydrocarbon group represented by R<sup>4</sup> and R<sup>5</sup> include alkylene groups such as ethylene, propylene, and butylene, and arylene groups such as phenylene and tolylene.

Examples of R<sup>f</sup> are shown below.

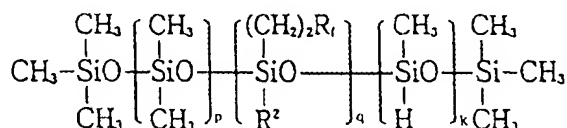




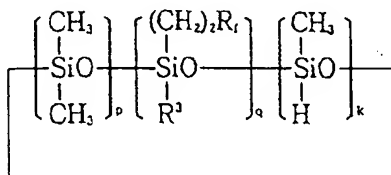
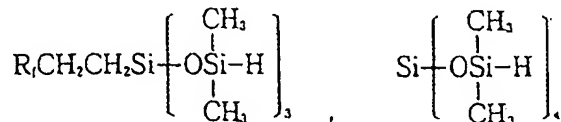
Among the fluorinated organosiloxanes of formula (1), those compatible with component (A) are preferred because uniformly cured products are expectable.

Component (A) or fluorinated organopolysiloxane and component (B) or cyclic fluorinated organosiloxanes are usually blended such that 0.01 to 30 parts, especially 0.05 to 15 parts by weight of component (B) is present per 100 parts by weight of component (A). In this respect, less than 0.01 part of component (B) tends to be too small to provide much adhesion whereas more than 30 parts of component (B) may sometimes detract from heat resistance and compression set.

Component (C) is an organohydrogenpolysiloxane having at least two hydrogen atoms attached to a silicon atom in a molecule. It serves as a crosslinking agent. Exemplary compounds are given below.



( $p \geq 0$ ,  $q \geq 0$ ,  $k > 0$ )



Copolymers consisting of  $(\text{CH}_3)_2\text{HSiO}_{1/2}$  and  $\text{SiO}_2$  units.

In the formulae,  $\text{R}^3$  and  $\text{R}_1$  are as previously defined, and letters  $p$ ,  $q$ , and  $k$  are numbers in the range  $p \geq 0$ ,  $q \geq 0$ , and  $k > 0$ .

Component (C) or organohydrogenpoly - siloxane is blended in the composition such that 0.5 to 5 mol, especially 0.5 to 3 mol of a hydrogen atom attached to a silicon atom (that is,  $\equiv \text{SiH}$  group) is available per mol of the aliphatic unsaturated group in the entire composition.

Component (D) is a platinum group metal compound serving as a curing catalyst. Examples include platinum black, chloroplatinic acid, complexes of chloroplatinic acid with alcohols, ethers, aldehydes, olefins (e.g., ethylene), vinylsilanes, and vinylsiloxanes, and platinum powder on supports such as alumina, silica and asbestos.

Preferably, component (D) or platinum group metal compound is added such that when calculated as metallic platinum, 1 to 500 ppm, especially 5 to 20 ppm of platinum is available based on the weight of component (A).

The compositions may contain further components e.g. selected from additives commonly used in conventional silicone rubbers. Such optional components include any of reinforcing fillers such as fumed silica,

precipitated silica, carbon powder, titanium dioxide, aluminum oxide, ground quartz, talc, sericite, and bentonite, fibrous fillers such as asbestos, glass fibers and organic fibers, dispersants such as diphenylsilane diol, hydroxyl-terminated dimethylpolysiloxane having a low degree of polymerization, and hexamethyldisilazane, heat resistance enhancers such as ferrous oxide, ferric oxide, cerium oxide, and iron octylate, and pigments. These additives may be used in conventional amounts.

The composition may be obtained by blending predetermined amounts of the various components, which may include such optional components, and uniformly kneading the mixture by any desired mixer.

The organopolysiloxane composition is typically cured by heating at a temperature of 100 to 200°C for about 30 seconds to about one hour, under atmospheric or increased pressure, to give a rubbery elastomer. If the composition is cured on a support, we find it can firmly adhere to the support.

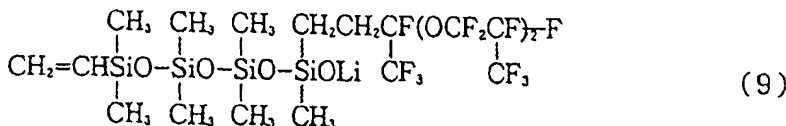
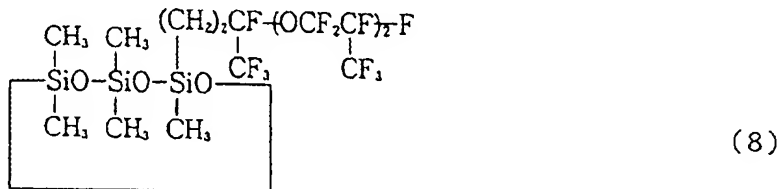
Insofar as the compositions have properties typical of fluorinated silicone rubber, and can cure to products having good adhesion, they may find use e.g. as packing, sealing and coating materials in electric and electronic fields.

#### EXAMPLE

Examples are given below by way of illustration and not by way of limitation. All parts are by weight.

#### Example 1 & Comparative Example 1

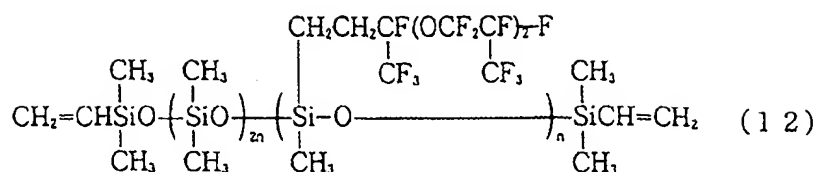
A 1 - liter separable flask equipped with a stirrer was charged with 1,000 grams (1.458 mol) of a cyclotrisiloxane of formula (8), 11.9 grams (0.0146 mol) of lithium silanolate of formula (9), and 1.0 grams of tetraglyme. In a nitrogen atmosphere, reaction was effected at 100°C for 5 hours.



The reaction mixture was cooled down. To the reaction mixture at 50°C were added 2.6 grams (0.0219 mol) of a compound of formula (10) and 6.1 grams (0.0328 mol) of a compound of formula (11). The reaction mixture was agitated for two hours for neutralization and terminal silylation.



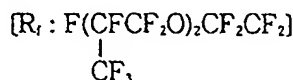
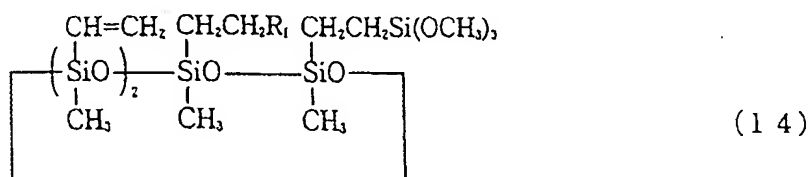
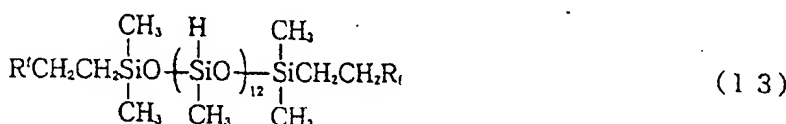
The reaction mixture was filtered and stripped, obtaining 938 grams of a polymer of formula (12) having a viscosity of 3,570 centistokes.



$$n \approx 100$$

In a kneader, 100 parts of the polymer of formula (12) and 15 parts of a fumed silica having a specific surface area of 300 m<sup>2</sup>/g were kneaded at 150°C for three hours.

To the mixture were added 1.8 grams of a compound of formula (13), 0.2 grams of carbon black, 0.1 gram of a toluene solution of a [(CH<sub>2</sub>=CHSi(CH<sub>3</sub>)<sub>2</sub>-O)-modified chloroplatinic acid catalyst, 0.4 grams of a 50% toluene solution of ethynylcyclohexanol, and 1.0 grams of a compound of formula (14) as a tackifier. They were mixed into a composition I.



Composition II was prepared by the same procedure as composition I except that the tackifier was omitted.

Each of compositions I and II was pressed at a temperature of 150°C and a pressure of 100 kg/cm<sup>2</sup> for 15 minutes into a sheet 2 mm thick. The sheet was heat treated at 150°C for one hour and then examined for rubber physical properties according to JIS K - 6301. The results are shown in Table 1.

A shearing test was carried out by fabricating a test sample as shown in FIG. 1. A pair of parallel plates 1 were joined together at one end through a sealing layer 2. An auxiliary plate was rested on the upper plate 1 and supported at an overhanging end by a spacer 3 on the lower plate 1 and at an overlapping end by another spacer via the upper plate 1. A weight 4 rested on the auxiliary plate above the sealing layer 2. In this condition, the plates 1 were pulled in opposite directions until the joint was broken. The plates 1 were made of glass, aluminum, iron or epoxy resin, the sealing layer 2 of cured composition I or II. In this way, the shearing adhesive force to glass, aluminum, iron and epoxy resin was measured. The results are shown in Table 2.

Table 1

	Example 1	Comparative Example 1
Composition	I	II
Hardness, JIS A	27	25
Elongation, %	300	320
Tensile strength, kgf/cm <sup>2</sup>	33	31

Table 2

5		Example 1		Comparative Example 1	
	Composition	I		II	
	Adherend	SAF	CF	SAF	CF
10	Glass	19.0	100	8.2	10
	Aluminum	16.5	100	7.2	0
15	Iron	20.5	100	6.5	0
	Epoxy resin	18.3	100	9.0	0

20 SAF: shearing adhesive force in kgf/cm<sup>2</sup>

CF: cohesive failure in %

25 As seen from Tables 1 and 2, the sheet of the composition embodying the present teachings has good rubber physical properties and is well adhesive to various supports.

The sheet of composition I was examined for swelling with various solvents (acetone, ethyl acetate, toluene, carbon tetrachloride and hexane). The percent volume and weight changes of the sheet were determined by immersing the sheet in a solvent at 20°C for 72 hours, measuring the volume and weight of the swollen sheet, and dividing the volume and weight of the swollen sheet by those of the initial sheet. For comparison purposes, dimethylsilicone rubber KE951 and fluorosilicone rubber FE 251 (trifluoropropylmethylsiloxane rubber type), both available from Shin-Etsu Chemical Co., Ltd., were similarly examined for solvent swelling. The results are shown in Table 3.

Table 3

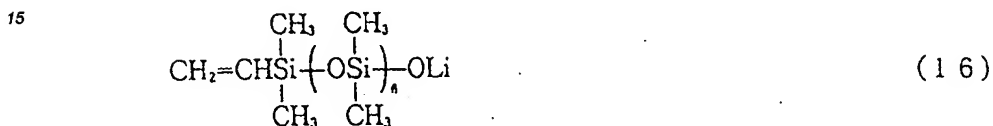
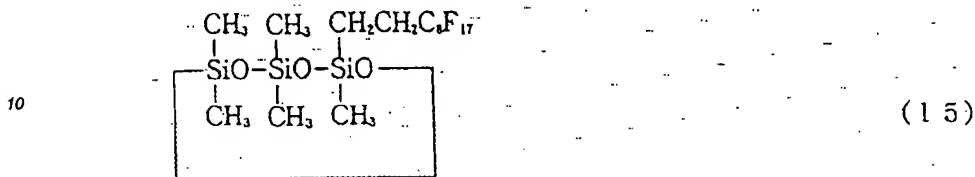
35			Example 1	Comparison	
	Solvent		Composition I	KE951	FE251
40	Acetone	volume change (%)	21	26	126
		weight change (%)	12	21	71
45	Ethyl acetate	volume change (%)	32	143	120
		weight change (%)	23	134	76
50	Toluene	volume change (%)	30	270	18
		weight change (%)	18	217	11
55	CCl <sub>4</sub>	volume change (%)	30	346	17
		weight change (%)	33	416	16
	Hexane	volume change (%)	19	186	10
		weight change (%)	9	119	2

As seen from Table 3, the sheet of the composition embodying the present teachings is well resistant against solvent swelling.



Example 2 & Comparative Example 2

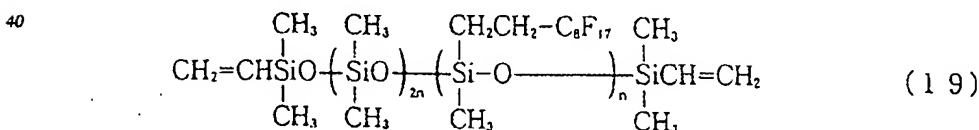
A 1 - liter separable flask equipped with a stirrer was charged with 1,000 grams (1.529 mol) of a cyclotri-  
 siloxane of formula (15), 7.5 grams (0.0153 mol) of lithium silanolate of average molecular formula (16), and  
 5 1.0 grams of tetraglyme. In a nitrogen atmosphere, reaction was effected at 100°C for 5 hours.



The reaction mixture was cooled down. To the reaction mixture at 50°C were added 2.8 grams (0.023 mol)  
 20 of a compound of formula (17) and 6.4 grams (0.035 mol) of a compound of formula (18). The reaction mixture  
 was agitated for two hours for neutralization and terminal silylation.



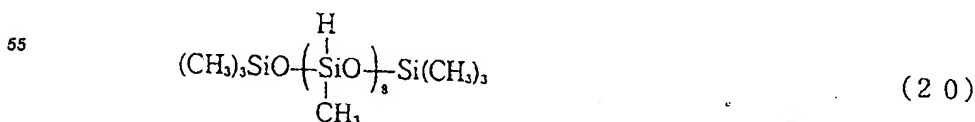
The reaction mixture was filtered and stripped, obtaining 911 grams of a polymer of formula (19) having  
 a viscosity of 10,510 centistokes.

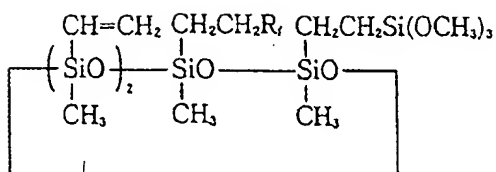


45  $n \approx 100$

In a kneader, 100 parts of the polymer of formula (19) and 15 parts of a fumed silica having a specific  
 surface area of 300 m<sup>2</sup>/g were kneaded at 150°C for three hours.

To the mixture were added 0.58 grams of a compound of formula (20), 0.2 grams of carbon black, 0.2 gram  
 50 of an alcohol solution of a chloroplatinic acid (platinum concentration 0.5%), 0.4 grams of a 50% toluene sol-  
 ution of ethynylcyclohexanol, and 1.0 grams of a compound of formula (21) as a tackifier. They were mixed  
 into a composition III.





(21)

(R<sub>f</sub> : C<sub>8</sub>F<sub>17</sub>)

Composition IV was prepared by the same procedure as composition III except that the tackifier was omitted.

As in Example 1, a sheet was press cured and a shearing test assembly was fabricated from each of composition III and IV. The measurements of physical properties are shown in Tables 4 and 5. The sheets were also examined for solvent swelling, with the results shown in Table 6.

Table 4

	Example 2	Comparative Example 2
Composition	III	IV
Hardness, JIS A	30	29
Elongation, %	350	370
Tensile strength, kgf/cm <sup>2</sup>	38	39

Table 5

	Example 2		Comparative Example 2	
Composition	III		IV	
Adherend	SAF	CF	SAF	CF
Glass	17.2	100	7.3	0
Aluminum	16.9	100	6.2	0
Iron	18.3	100	5.9	0
Epoxy resin	17.5	100	9.2	10

SAF: shearing adhesive force in kgf/cm<sup>2</sup>

CF: cohesive failure in %

Table 6

		Example 2
Solvent		Composition III
Acetone	volume change (%)	16
	weight change (%)	8
Ethyl acetate	volume change (%)	25
	weight change (%)	14
Toluene	volume change (%)	23
	weight change (%)	12
CCl <sub>4</sub>	volume change (%)	25
	weight change (%)	23
Hexane	volume change (%)	16
	weight change (%)	8

As seen from Tables 4, 5, and 6, the composition embodying the present teachings had good rubber physical properties, good adhesion to various supports, and resistance against solvent swelling.

Although some preferred embodiments have been described, many modifications and variations may be made thereto in the light of the above teachings. It is therefore to be understood that within the scope of the concepts disclosed herein, the compositions may be otherwise than as specifically described in the examples.

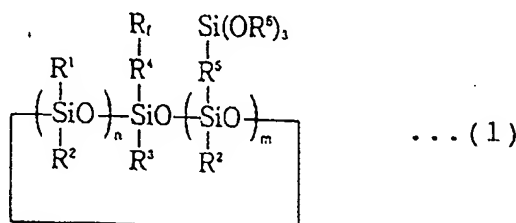
It will therefore be understood that, in one general aspect, the present invention provides a fluorinated organopolysiloxane composition, hydrosilation-curable in the presence of a catalyst to give a fluorinated silicone elastomer, characterised by the inclusion of a cyclic fluorinated organosiloxane having side groups -R<sup>1</sup>, R<sup>4</sup>-R<sub>f</sub> and -R<sup>5</sup>-Si(OR<sup>6</sup>)<sub>3</sub> (as explained above) on silicon atoms, to give adhesive capability to the composition.

### Claims

1. An organopolysiloxane composition comprising

(A) a linear fluorinated organopoly-siloxane having in a molecule at least two monovalent hydrocarbon groups each having an aliphatic unsaturated bond attached to a silicon atom, and further having at least one group selected from monovalent perfluoroalkyl and monovalent perfluoropolyether groups attached to a silicon atom,

(B) a cyclic fluorinated organosiloxane of the general formula (1):



wherein R<sup>1</sup> is a substituted or unsubstituted monovalent hydrocarbon group having an aliphatic unsaturated bond, R<sup>2</sup>, R<sup>3</sup> and R<sup>6</sup> are independently selected from substituted or unsubstituted monovalent hydrocarbon groups free of an aliphatic unsaturated bond, R<sup>4</sup> and R<sup>5</sup> are independently selected from substituted or unsubstituted divalent hydrocarbon groups free of an aliphatic unsaturated bond, R<sub>f</sub> is a monovalent perfluoropolyether group or monovalent perfluoroalkyl group, and letters m and n are

numbers satisfying  $n + m = 3$ ,  $n \geq 1$ , and  $m \geq 1$ , as a tackifier,

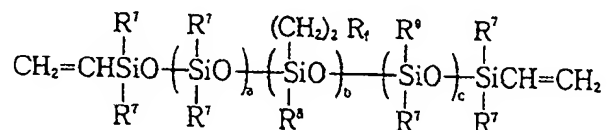
(C) an organohydrogenpolysiloxane having at least two hydrogen atoms attached to a silicon atom in a molecule, and

(D) a platinum group metal catalyst.

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2. An organopolysiloxane composition according to claim 1 wherein fluorinated organopolysiloxane (A) is a polymer of the general formula (7):

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(7)

wherein  $\text{R}^7$  and  $\text{R}^8$  are independently selected from substituted or unsubstituted monovalent hydrocarbon groups free of an aliphatic unsaturated bond,  $\text{R}^9$  is a monovalent hydrocarbon group having an aliphatic unsaturated bond,  $\text{R}_f$  is a monovalent perfluoropolyether group or monovalent perfluoroalkyl group, letters a, b and c are integers in the range: a = 2 to 3,000, b = 1 to 1,000, and c = 0 to 50.

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3. An organopolysiloxane composition according to claim 1 which contains  
 100 parts by weight of component (A),  
 0.01 to 30 parts by weight of component (B),  
 component (C) in an amount to provide 0.5 to 5 mol of SiH group per mol of the aliphatic unsaturated group in the entire composition and  
 1 to 500 ppm calculated as platinum of component (D) based on the weight of component (A).
4. A method comprising the preparation of an organopolysiloxane composition according to any one of claims 1 to 3, by blending of the ingredients thereof.

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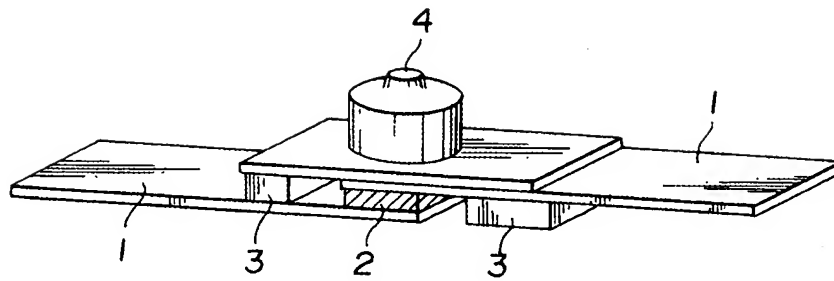
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**FIG.1**





European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number  
EP 94 30 1634

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	EP-A-0 393 984 (SHIN-ETSU) * page 7; claim 1 *	1	C08L83/08
A	US-A-4 355 149 (TORAY) * claim 1; example 1 *	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C08L C09D C08G
The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Examiner
THE HAGUE		30 June 1994	Lentz, J
<p><b>CATEGORY OF CITED DOCUMENTS</b></p> <p>X : particularly relevant if taken alone  Y : particularly relevant if combined with another document of the same category  A : technological background  O : non-written disclosure  P : intermediate document</p> <p>T : theory or principle underlying the invention  E : earlier patent document, but published on, or after the filing date  D : document cited in the application  L : document cited for other reasons  &amp; : member of the same patent family, corresponding document</p>			

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